A Unified Methodology for Modeling Hysteresis in Ferroelectric, Ferromagnetic and Ferroelastic Materials

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Abstract

This investigation focuses on the development of unified techniques for mathematically modeling the hysteresis and constitutive nonlinearities inherent to ferroelectric, ferromagnetic and ferroelastic materials at moderate to high drive levels. Motivating materials include piezoceramics, relaxor ferroelectrics, magnetostrictives and shape memory alloys, but the modeling approach is sufficiently general to include a large variety of ferroic compounds. The nonlinear and hysteretic behavior of these materials can be attributed to their underlying domain structure and this common ferroic framework is utilized to construct unified constitutive models for the materials. These models are constructed in two steps. In the first, thermodynamic principles are employed to quantify the anhysteretic behavior which would result in the absence of inclusions in the material. In the second step, energy relations are employed to quantify the irreversible and reversible motion of domains walls about pinning sites in the material. The resulting models are formulated as low-order ordinary differential equations. The performance and behavior of the models are illustrated for piezoceramic, magnetostrictive and shape memory compounds.

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1 Introduction

Piezoceramic, relaxor ferroelectric, magnetostrictive and shape memory compounds all exhibit varying degrees of hysteresis and constitutive nonlinearities which depend upon the drive levels and operating conditions being employed. While the linearity of the material's response can for certain regimes be improved through the design of the drive electronics (e.g., current control for piezoceramic devices) or feedback mechanisms, the quantification of mechanisms producing the nonlinearities and hysteresis will ultimately improve the performance capabilities of actuators and sensors which employ these materials. The modeling of mechanisms which govern the constitutive behavior of the materials also facilitates the development of new compounds which have improved performance. Finally, the performance of control algorithms used to regulate the behavior of actuators or sensors that utilize these materials will be improved through the development of accurate material models.

In this paper, we discuss a unified modeling strategy for ferroelectric, ferromagnetic and ferroelastic materials based on the common ferroic nature of the compounds. Specifically, we consider ferroic materials to be those characterized by domain walls which can be reoriented by external fields or stresses [36], and it is the associated domain switching and domain wall mechanisms common to these materials that we exploit for model development. While mechanisms which produce these phenomena differ at the microscopic level, the energy mechanisms at quasi-static levels are sufficiently similar to permit the development of unified theories. The theory presented here is developed in two steps. In the first, thermodynamic principles are employed to quantify the anhysteretic (hysteresis-free) behavior that would occur for unimpeded domain wall movement. Secondly, the energy required to reorient domains is employed to quantify reversible and irreversible effects due to domain wall bending and translation across pinning sites. The resulting model quantifies the polarization, magnetization, or strains respectively exhibited by ferroelectric, ferromagnetic or ferroelastic materials in response to input fields or stresses. By utilizing effective material parameters obtained through macroscopic averaging of microscopic material properties, the resulting models are formulated as low-order ODE which facilitates material characterization and subsequent control design.

The methodology developed here complements certain existing techniques for developing unified models for the considered materials. Because the mechanisms which govern the domain dynamics and resulting nonlinearities and hysteresis differ at the microscopic level, the unified models are necessarily phenomenological or quasi-macroscopic in nature. The former category includes a variety of Preisach models which characterize static hysteresis in these compounds [1, 2, 3, 4, 19, 20, 37, 39, 52, 56]. The advantage of this approach lies in the mathematical structure which enforces criteria such as minor loop closure when the underlying physical mechanisms are poorly understood or difficult to quantify. However, because the coefficients in Preisach models are nonphysical, it is difficult to utilize known physics to simplify model development or to facilitate parameter updating to accommodate changing operating conditions. A second unified approach for quantifying the nonlinear and hysteretic behavior of certain active materials has been developed by Soukhojak and Chiang [51]. Based on phenomenological principles, this approach provides a rheological model quantifying the time and frequency-dependent behavior of a variety of ferroelectric, ferromagnetic or ferroelastic compounds. Finally, we note that several unified models have been developed which quantify constitutive nonlinearities in electrostrictive and magnetostrictive materials in the absence of hysteresis.

Models characterizing the anhysteretic behavior of ferroic materials are summarized in Section 2 while domain wall losses are incorporated in Section 3 to provide a unified modeling strategy for the compounds. Examples illustrating the performance of the model for characterizing the polarization, magnetization and strains in piezoceramic, magnetostrictive and shape memory compounds are provided in Section 4.

1

2   Anhysteretic Relations

To provide a common framework for quantifying the anhysteretic behavior of ferroelectric, ferromagnetic and ferroelastic materials, we consider first the formulation of appropriate free energy relations which are assumed to be functions of the absolute temperature T and an order parameter e. The latter is chosen to differentiate between phases in the materials, and for ferroelectric, ferromagnetic and ferroelastic compounds, e is respectively specified to be the polarization P, magnetization M, or strain ε. For a summary of the thermodynamic relations and theory of phase transitions pertinent to this discussion, we refer the reader to [5, 53].

Throughout this discussion, we let F denote the Helmholtz free energy and let $\widetilde{\varphi}$ denote external fields that are thermodynamically conjugate to e. For the order parameters P, M and ε, appropriate choices for the external field are $\widetilde{\varphi}$ are the electric field E, magnetic field H or stress σ.

In the absence of applied fields, thermodynamic equilibria are determined by minimizing F with respect to e which, under the assumption of differentiability, yields the condition

$$\varphi(e,T) \equiv \frac{\partial F}{\partial e} = 0 \tag{1}$$

where φ denotes the energetic response of the system. For systems that are subjected to external fields, the total free energy is

$$F_{\widetilde{\varphi}}(e,T) = F(e,T) - \widetilde{\varphi}e \tag{2}$$

which yields the equilibrium condition

$$\varphi(e,T) = \widetilde{\varphi} \,. \tag{3}$$

In order to employ the equilibrium conditions (1) or (3) to determine appropriate constitutive relations, it is necessary to specify the free energy relations F or $F_{\widetilde{\varphi}}$. We consider two techniques through which this can be accomplished: (i) truncated power series expansions, and (ii) Boltzmann relations. We focus first on the formulation of general energy relations and then provide specific examples for the classes of materials under consideration.

In the theory of Landau and Devonshire, it is assumed that the free energy can be expressed as a power series

$$F(e,T) = \sum_{i=0}^{\infty} F_i(T)e^i \tag{4}$$

in terms of the order parameter [5, 14, 30, 53]. The coefficients $F_i(t)$ are chosen to enforce phenomenological properties of first or second-order phase transitions and the series are typically truncated after $i = 6$.

For materials which exhibit second-order phase transitions, the assumption that $F_6(T) \equiv 0$ yields the Landau form

$$F(e,T) = F_0(T) + \alpha_1(T - T_c)e^2 + \alpha_2 e^4 \tag{5}$$

where T_c denotes the critical temperature and α_1, α_2 are positive constants. The term $F_0(T)$ incorporates all contributions which are not dependent on the order e. As depicted in Figure 1a, this yields a single well potential for temperatures $T > T_c$ and a double well potential potential for $T < T_c$. Hence the thermodynamic potential produces single-valued anhysteretic curves above transition temperature and multi-valued curves below this temperature (see Figure 1b, 1c). The free energy expression (5) is appropriate for ferromagnetic materials as well as certain ferroelectric compounds (e.g., Rochelle salt and KH_2PO_4 – see Devonshire [14]).

Alternatively, the functional

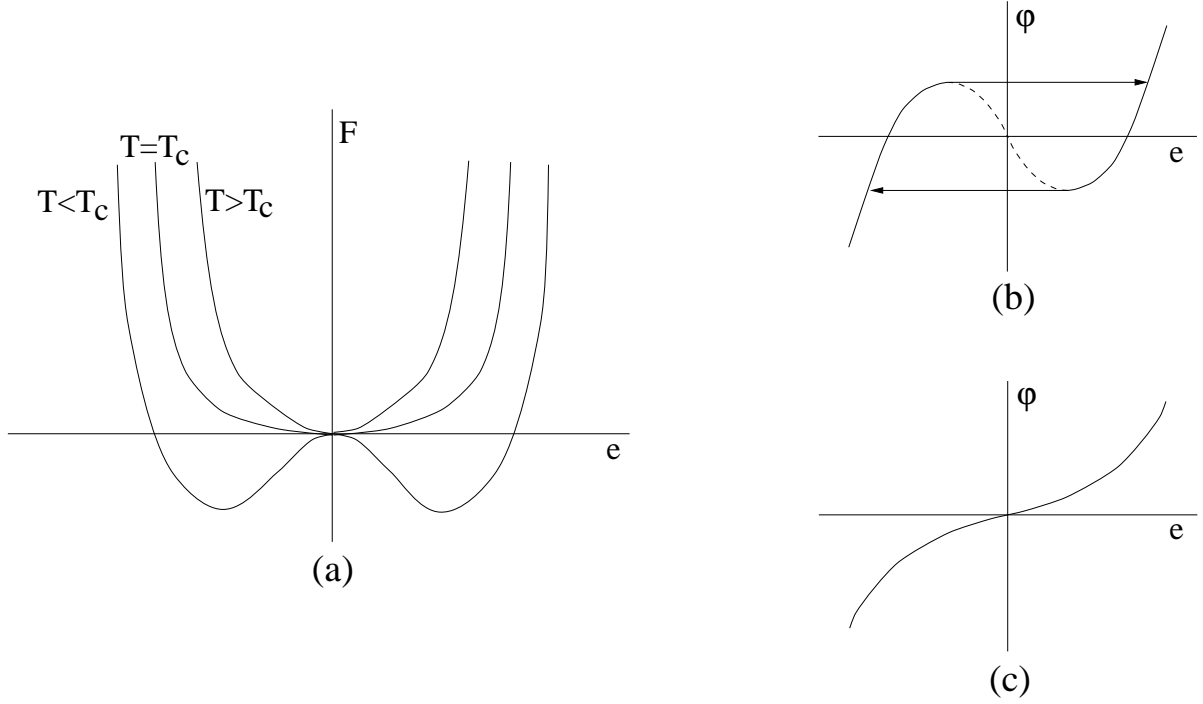$$F(e,T) = F_0(T) + \alpha_1(T - T_c)e^2 - \alpha_2 e^4 + \alpha_3 e^6, \tag{6}$$

2

Figure 1. (a) Landau free energy F for materials exhibiting second-order phase transitions; (b) Relation between the order e and energy response function φ for $T < T_c$, and (c) $T \geq T_c$.

where α_1, α_2 and α_3 are positive, can be employed for materials which exhibit first-order phase transitions (e.g., shape memory alloys and certain ferroelectric compounds including BaTi). As detailed in [5, 14, 29] and depicted in Figure 2, this functional can have up to three minima. The central minima at $e = 0$ represents the high temperature phase while the lateral twin minima occur at low temperatures. Details regarding the resulting anhysteretic and hysteretic relations between e and φ can be found in [5].

It is noted that in the theories of Landau and Devonshire, non-local effects are neglected when employing functionals of the form $F(e, T)$. The incorporation of non-local spatial effects can be accomplished by using the Landau-Ginzburg functional

$$\mathcal{F}[e, T] = \int_{\Omega} \left[F(e(\mathbf{x}), T) + \frac{1}{2}\mu(e(\mathbf{x}), T)|\nabla e(\mathbf{x})|^2 \right] d\mathbf{x} \tag{7}$$

where μ is positive function of e and T and the order parameter now exhibits spatial variation. It is observed that the inclusion of the gradient term serves to act as a regularization term in the expression. As discussed in [40], it is necessary to employ some form of Landau-Ginzburg functional when modeling domain walls in a thermodynamic framework to accommodate the inherent spatial variations which occur in these transition regions. The establishment of well-posedness criteria for models employing Landau-Ginzburg type functions can be found in [5].

A second method for specifying free energy functionals F is to employ Boltzmann principles to balance the thermal energy with the electrostatic, magnetostatic, or elastic energies of the constituent compounds. Details regarding this modeling approach will be provided in subsequent discussion when the specific materials are addressed.
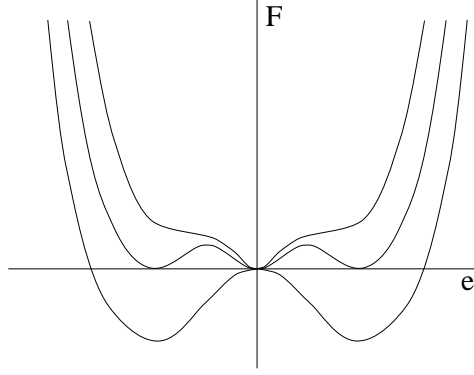
Figure 2. (a) Devonshire free energy F for materials exhibiting first-order phase transitions.

2.1 Ferroelectric Materials

As noted previously, ferroelectric materials can exhibit either first or second-order transitions between the paraelectric phase, which occurs above the Curie temperature, and the ferroelectric phase that occurs below this temperature. Additionally, materials such as relaxor ferroelectric compounds exhibit superparaelectric states which produce a broad transition between the ferroelectric and paraelectric states.

For materials that exhibit second-order phase transitions and linear coupling between the elastic and electric variables, we consider the free energy expression

$$F(\varepsilon, P, T) = \frac{1}{2}c^P\varepsilon^2 - \eta\varepsilon P + F_0(T) + \alpha_1(T - T_c)P^2 + \alpha_2 P^4 \,. \tag{8}$$

Here c^P denotes the Young's modulus at constant polarization and η is a coupling coefficient. Enforcement of the equilibrium condition (3) yields the stress and field relations

$$\sigma = \left[\frac{\partial F}{\partial \varepsilon}\right]_{P,T} \quad , \quad E = \left[\frac{\partial F}{\partial P}\right]_{\varepsilon,T} \tag{9}$$

which provides the constitutive relation

$$\sigma = c^P\varepsilon - \gamma P$$
$$E = -\gamma\varepsilon + 2\alpha_1(T - T_c)P + 4\alpha_2 P^3 \,. \tag{10}$$

The relation (10) provides a constitutive model for piezoelectric materials that includes a form of temperature-dependence along with constitutive nonlinearities due to saturation. When quantifying the anhysteretic polarization due to an applied field, it is advantageous to express the polarization P quantified by (10) as a function of the input field. As illustrated in Figure 3, this relation incorporates a form of hysteresis due to the double well potential at temperatures below the Curie temperature. However, the anhysteretic model does not incorporate domain wall losses which involve irreversible processes of the type detailed in [33]. Mechanisms for addressing these effects will be discussed in Section 3.

The free energy expression (8) and resulting constitutive expressions must be modified for electrostrictive materials to accommodate the quadratic dependence of the strain on the polarization. An appropriate free energy functional for this case is

$$F(\varepsilon, P, T) = \frac{1}{2}c^P\varepsilon^2 - \gamma\varepsilon P^2 + F_0(T) + \alpha_1(T - T_c)P^2 + \alpha_2 P^4 \tag{11}$$
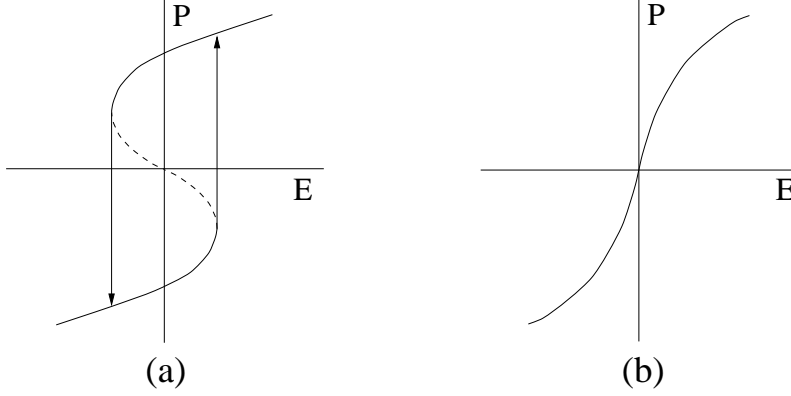
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Figure 3. Relation between P and E quantified by (10) for (a) $T < T_c$ and (b) $T > T_c$.

which yields the constitutive relations

$$
\begin{aligned}
\sigma &= c^P e - \gamma P^2 \\
E &= -2\gamma\varepsilon P + 2\alpha_1(T - T_c)P + 4\alpha_2 P^3 \,.
\end{aligned}
\tag{12}
$$

Alternative free energy expressions can be obtained from Boltzmann principles. To illustrate, we summarize a model employed in [22, 23, 24] for electrostrictive materials. As detailed in [24], the internal energy due to short range interactions can be quantified by

$$
U = \frac{\Phi_0 N}{4V}\left[1 - (P/P_s)^2\right]
\tag{13}
$$

where N is the number of cells, V is the volume of the lattice, Φ_0 is the energy required to convert the spin at a single site, and $P_s = Np_0/V$ is the saturation polarization. Here p_0 denotes the dipole moment of a single cell within the lattice. The application of statistical mechanics then yields the entropy expression

$$
S = \frac{k_b N}{V}\left[\ln(2) - \frac{1 + P/P_s}{2}\ln(1 + P/P_s) - \frac{1 - P/P_s}{2}\ln(1 - P/P_s)\right]
\tag{14}
$$

where k_b is Boltzmann's constant [21]. The employment of (13) and (14) in the free energy relation

$$
F = U - TS
$$

and inclusion of electromechanical effects then yields the temperature-dependent free energy expression

$$
\begin{aligned}
F(\varepsilon, P, T) = {}&\frac{1}{2}c^P\varepsilon^2 - \gamma\varepsilon P^2 + \frac{E_h P_s}{2}\left[1 - (P/P_s)^2\right] \\
&+ \frac{E_h T}{2T_c}\left[P\ln\left(\frac{P + P_s}{P_s - P}\right) + P_s\ln\left(1 - (P/P_s)^2\right)\right]
\end{aligned}
\tag{15}
$$

employed in [22, 23, 24] for electrostrictive materials. Here $E_h = \Phi_0/(2p_0)$ denotes a material parameter which is typically estimated through a least squares fit to data while $T_c = \Phi_0/(2P_s k_B)$ denotes the Curie temperature for the material. An analogous expression can be derived for piezoelectric materials by considering a coupling relation which is linear with respect to the polarization.

To obtain constitutive relations, the constraints (9) resulting from (3) are invoked to yield

$$\sigma = c^P \varepsilon - \gamma P^2$$
$$E = -2\gamma\varepsilon P - \frac{E_h}{P_s}P + \frac{E_h T}{T_c}\text{arctanh}(P/P_s) \tag{16}$$

To compare the constitutive relations (12) and (16), the hyperbolic function is represented by its Taylor series to yield

$$
\begin{aligned}
E &= -2\gamma\varepsilon P - \frac{E_h}{P_s}P + \frac{E_h T}{T_c}\left[\left(\frac{P}{P_s}\right) + \frac{1}{3}\left(\frac{P}{P_s}\right)^3 + \frac{1}{5}\left(\frac{P}{P_s}\right)^5 + \cdots\right] \\
&= -2\gamma\varepsilon P + \frac{E_h}{T_c P_s}(T - T_c)P + \frac{E_h T}{3 T_c P_s^3}P^3 + \frac{E_h T}{5 T_c P_s^5}P^5 + \cdots.
\end{aligned}
$$

In this form, it is observed that (12) is a special case of (16) in which the parameters α_1 and α_2 are given by

$$\alpha_1 = \frac{E_h}{2 T_c P_s} \quad , \quad \alpha_2(T) = \frac{E_h T}{12 T_c P_s^3}.$$

The field relations in any of the constitutive expressions (10), (12) or (16) can be employed as models for the anhysteretic polarization. However the expression (16) derived from Boltzmann principles is typically employed for two reasons: (i) it can be inverted to accommodate field inputs and (ii) by employing an alternative derivation from statistical mechanics, additional anhysteretic models can be derived. To illustrate the first property, we consider for simplicity the case where $\varepsilon = 0$. Reformulation of (16) then yields

$$P_{an} = P_s\tanh(E_e/a) \tag{17}$$

where

$$E_e = E + \alpha P \tag{18}$$

denotes the effective field generated in the material and $\alpha = E_h/P_s$ and $a(T) = E_h T/T_c$. The Ising spin relation (17) provides one of the models used to characterize the anhysteretic polarization in ferroelectric materials.

An alternative derivation of (17) illustrates that the Ising relation is actually one member of a family of models which can be employed to quantify the anhysteretic polarization in ferroelectric materials or the anhysteretic magnetization in ferromagnetic materials. As detailed in [44], these models can be derived by employing Boltzmann statistics to express the probability that dipoles occupy certain energy states. For noninteracting cells, the probability that a dipole occupies the energy state $\mathcal{E}$ can be expressed as

$$\mu(\mathcal{E}) = Ce^{-\mathcal{E}/k_B T} \tag{19}$$

where $k_B T$ is the thermal energy and the parameter C is chosen to ensure that integration over all possible configurations yields the total number of moments per unit volume N. For a dipole $\mathbf{p}$ in an electric field $\mathbf{E}$, the potential energy is

$$\mathcal{E} = -\mathbf{p} \cdot \mathbf{E}. \tag{20}$$

Under the assumption that dipoles can orient only in the direction of the applied field or opposite to it, integration and incorporation of neighboring effects yields the macroscopic Ising spin model

$$P = P_s\tanh(E_e/a) \tag{21}$$

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whereas the Langevin model

$$P = P_s \left[\coth\left(\frac{E_e}{a}\right) - \left(\frac{a}{E_e}\right) \right] \tag{22}$$

derives from the assumption that the cell's orientation can be in any direction. In both cases, effective field E_e is specified by (18). The Ising model has been primarily used when characterizing ferroelectric materials whereas the Langevin model is widely employed in ferromagnetic applications. We note, however, that the Langevin model cannot be be analytically inverted to provide constitutive relations analogous to those derived from corresponding free energy relations. Finally, additional models derived in this manner are discussed in [44] while further details regarding the properties of anhysteretic models for relaxor ferroelectric materials are provided in [38].

2.2 Ferromagnetic Materials

While the mechanisms which produce dipole switching and domain wall movement in ferroelectric and ferromagnetic materials differ quite significantly at the molecular level, the resulting qualitative phenomena are sufficiently similar to permit the use of analogous methodologies when developing anhysteretic models and quasi-macroscopic domain wall relations. Consequently, either power series representations or Boltzmann arguments can be used to drive free energy expressions, constitutive relations and anhysteretic models for ferromagnetic materials which are analogous to those developed for ferroelectric compounds.

To illustrate, we consider the development of a free energy relation from Boltzmann principles. As detailed in [24], the quantification of the energy required to reorient spins yields a relation for the internal energy which can be combined with classical entropy relations to yield the inelastic Helmholtz relation

$$F(M, T) = \frac{E_h M_s}{2}\left[1 - (M/M_s)^2\right] + \frac{E_h T}{2T_c}\left[M\log\left(\frac{M + M_s}{M_s - M}\right) + M_s \log\left(1 - (M/M_s)^2\right)\right]$$

where M_s denotes the saturation magnetization for a given material and E_h again denotes a coupling field. As detailed in [9, page 343], [10, page 279], magnetostrictive materials exhibit a quadratic relation between the magnetization and strain. Hence an appropriate elastic free energy expression is

$$F(\varepsilon, M, T) = \frac{1}{2}c^M \varepsilon^2 - \gamma\varepsilon M^2 + \frac{E_h M_s}{2}\left[1 - (M/M_s)^2\right]$$
$$+ \frac{E_h T}{2T_c}\left[M\ln\left(\frac{M + M_s}{M_s - M}\right) + M_s \ln\left(1 - (M/M_s)^2\right)\right] \tag{23}$$

where γ denotes a coupling coefficient. The enforcement of (3) yields the equilibrium constraints

$$\sigma = \left[\frac{\partial F}{\partial \varepsilon}\right]_{M,T} \quad , \quad H = \left[\frac{\partial F}{\partial M}\right]_{\varepsilon,T}$$

and resulting constitutive relations

$$\sigma = c^M \varepsilon - \gamma M^2$$
$$H = -2\gamma\varepsilon M - \alpha M + \frac{E_h T}{T_c}\text{arctanh}(M/M_s) \tag{24}$$

where $\alpha = E_h/M_s$. The combined relations are analogous to the constitutive relations derived in [15] and the stress relation is precisely that employed in [13]. Further details regarding the comparison between linear constitutive relations for ferromagnetic and ferroelectric materials are provide in [32].

In the absence of applied strains, the inversion of the field relation yields the anhysteretic model

$$M = M_s \tanh(H_e/a) \, . \tag{25}$$

The effective field is specified by

$$H_e = H + \alpha M \tag{26}$$

where for fixed T and M, $a = E_h T/T_c$.

We note that as with ferroelectric materials, the Ising anhysteretic model can also be derived by computing the probability of attaining various dipole configurations (and hence corresponding energy states) due to an applied effective field. While this approach is equivalent to that employed when quantifying the energy required to reorient dipoles when computing the internal energy, it provides the advantage of yielding a family of anhysteretic models which differ according to the underlying assumptions made regarding possible dipole and grain orientations. The Ising relation (25) results from the assumption that dipoles can orient only in the direction of, or opposite to, the applied field. As detailed in [9, 27], the less restrictive assumption that dipoles can orient uniformly yields the Langevin model

$$M = M_s \left[\coth \left(\frac{H_e}{a} \right) - \left(\frac{a}{H_e} \right) \right] \tag{27}$$

which is more commonly employed in magnetic applications. It should be noted that the use of the Langevin model for ferromagnetic materials having mean field interactions should be interpreted as phenomenological in nature since the theory is derived for paramagnetic materials in which magnetic moments are localized in atomic sites [27, page 187]. However, it has been demonstrated that the Langevin relation adequately quantifies the anhysteretic behavior of a number of metals and rare earth compounds in spite of these theoretical limitations.

2.3   Ferroelastic Materials

The phenomenological behavior of ferroelastic materials is in general very complicated due to the complex and highly coupled behavior of the constituent materials, the magnitude of generated strains, and the constitutive nonlinearities inherent to the materials. However, initial anhysteretic models have been developed using the previously described techniques and subsequently employed as kernels in nonlinear hysteresis models.

We consider first ferroelastic materials, such as shape memory alloys, which exhibit first-order phase transitions and we illustrate with a one-dimensional model developed by Falk for shape memory alloys [16, 17]. As summarized in [5], this model employs the total Landau-Ginzburg free energy

$$\mathcal{F}(\varepsilon, \varepsilon_x, T) = F(\varepsilon, T) + \frac{\mu}{2} \varepsilon_x^2 \tag{28}$$

where ε_x denotes the gradient of the strain and the local free energy

$$F(\varepsilon, T) = F_0(T) + \alpha_1 (T - T_c) \varepsilon^2 - \alpha_2 \varepsilon^4 + \alpha_3 \varepsilon^6 \tag{29}$$

results from the truncation of (1). The parameters α_1, α_2 and α_3 are assumed to be positive to guarantee first-order behavior. The inclusion of the quadratic strain term incorporates nonlocal effects such as interfacial energies. A comparison of the local free energy (29) with that employed in (8) and (11) for ferroelectric materials illustrates that two have the same form with differences due only to the choice of order parameter and the sign of α_2 which is chosen to ensure first or second-order phase transitions.

For materials subjected to an external stress σ, the total free energy relation (2) can be employed to yield

$$F(\varepsilon, T, \sigma) = \alpha_1 (T - T_c)\varepsilon^2 - \alpha_2 \varepsilon^4 + \alpha_3 \varepsilon^6 - \sigma \varepsilon \,. \tag{30}$$

The enforcement of the equilibrium condition $\frac{\partial F}{\partial \varepsilon} = 0$ yields the constitutive relation

$$\sigma = 2\alpha_1 (T - T_c)\varepsilon - 4\alpha_2 \varepsilon^3 + 6\alpha_3 \varepsilon^5 \tag{31}$$

which implicity quantifies the strains that result from an applied stress.

The analogous expressions

$$F(\varepsilon, T, \sigma) = \alpha_1 (T - T_c)\varepsilon^2 + \alpha_2 \varepsilon^4 + \alpha_3 \varepsilon^6 - \sigma \varepsilon \,. \tag{32}$$

and

$$\sigma = 2\alpha_1 (T - T_c)\varepsilon + 4\alpha_2 \varepsilon^3 + 6\alpha_3 \varepsilon^5 \tag{33}$$

can be employed for the free energy and resulting stress in materials which undergo second-order phase transitions. The constitutive relation (33), employed in conjunction with the effective stress

$$\sigma_e = \sigma + \alpha \varepsilon \tag{34}$$

has been employed by Tuszyński et al. [55] to provide a model for anhysteretic strains generated in LiCsSO$_4$. The inclusion of the component $\alpha \varepsilon$ incorporates mean field contributions in the manner exhibited by αP and αM in the effective field relations (18) and (26). Details regarding the role of this term in general ferroelastic compounds are provided in [40]. For the model described in [55], the temperature $T = \widehat{T}$ is held fixed, and constants are given by

$$\alpha_1 = \frac{c_{2s}}{\widehat{T} - T_c} \quad , \quad \alpha_2 = \frac{c_{3ms}^2}{c_{3mm}} - 4c_{4s} \quad , \quad \alpha_3 = 6c_{6s} - \frac{3c_{3ms}^2 c_{4mms}}{2c_{mm}^2}$$

where c_{2s} and c_{mm} are second-order elastic coefficients, c_{3ms} are third-order coefficients, c_{4s} and c_{4mms} are fourth-order elastic coefficients and c_{6s} is a sixth-order elastic coefficient. We note that when models of the form (31) or (33) are employed for general material characterization, the coefficients α_1, α_2 and α_3 are treated as parameters which must be estimated through a least squares fit to data.

The free energy expression (30) has been employed for LiCsSO$_4$ with the relation (31) used to quantify anhysteretic strains [55]. As will be detailed in the next section, the resulting anhysteretic strains can then be employed as the minimal energy component in hysteresis models which quantify losses due to domain wall pinning.

2.4   Anhysteretic Models

We summarize here the models developed in previous sections for the anhysteretic polarization, magnatization and strain. In each case, the models incorporate certain constitutive nonlinearities along with a form of hysteresis consistent with domain switching as modeled by double well potential theory. Hence the models can be interpreted as representing thermodynamic equilibria for elastic, magnetic and electric regimes. The models do not incorporate the reversible or irreversible effects of domain wall motion, and the inclusion of these mechanisms is addressed in the next section.

Note that in the respective definitions, the notation e_{an} denotes the anhysteretic value whereas e is the prevailing value of the order parameter $e = P, M, \varepsilon$. For the electric field relation, we include the case deriving from quadratic coupling between the strain and polarization (electrostriction) and simply note that for materials which exhibit linear coupling (e.g., piezoceramics), analogous field relations can be derived.

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Polarization: $E_e = E + \alpha P$

$$E_e = -2\gamma\varepsilon P + 2\alpha_1(T - T_c)P_{an} + 4\alpha_2 P_{an}^3 \qquad \text{(Second-Order Phase Transition)}$$

$$P_{an} = P_s \tanh\left(\frac{E_e}{a}\right) \qquad\qquad\qquad \text{(Ising Model)}$$

$$P_{an} = P_s\left[\coth\left(\frac{E_e}{a}\right) - \left(\frac{a}{E_e}\right)\right] \qquad\qquad \text{(Langevin Model)}$$

(35)

Magnetization: $H_e = H + \alpha M$

$$M_{an} = M_s \tanh\left(\frac{H_e}{a}\right) \qquad\qquad\qquad \text{(Ising Model)}$$

$$M_{an} = M_s\left[\coth\left(\frac{H_e}{a}\right) - \left(\frac{a}{H_e}\right)\right] \qquad\qquad \text{(Langevin Model)}$$

(36)

Strain: $\sigma_e = \sigma + \alpha\varepsilon$

$$\sigma_e = 2\alpha_1(T - T_c)\varepsilon_{an} + 4\alpha_2\varepsilon_{an}^3 + 6\alpha_3\varepsilon_{an}^5 \qquad \text{(Second-Order Phase Transition)}$$

$$\sigma_e = 2\alpha_1(T - T_c)\varepsilon_{an} - 4\alpha_2\varepsilon_{an}^3 + 6\alpha_3\varepsilon_{an}^5 \qquad \text{(First-Order Phase Transition)}$$

(37)

3   Domain Wall Losses

The anhysteretic models developed in the previous section quantify domain switching in ferroelectric, ferromagnetic and ferroelastic materials under the assumption that domain walls are unimpeded and hence conservative. These models quantify constitutive nonlinearities and at low temperatures, yield hysteresis curves characteristic of double-well potential theory. As observed in Figure 3, however, these curves exhibit a jump between branches due to the instability of the central equilibrium at $e = 0$. This produces transitions that are more abrupt than typically observed for physical materials. Two mechanisms which prevent such steep transitions are the bending and translation of domain walls pinned at inclusions in the materials. Domain wall bending produces reversible changes in the order parameter, in a manner analogous to surface tension in a bubble, while the translation of domain walls produces irreversible effects due to movement across minima in the potential energy for the system.

Models quantifying both mechanisms are outlined in this section. To clarify the presentation, we will consider individually the models for ferroelectric, ferromagnetic and ferroelastic materials and then pose them in a common framework and notation at the end of the section. Examples illustrating the behavior and performance of the models are provided in the next section.

3.1   Ferroelectric Materials

As illustrated in [26, 34], the behavior of ferroelectric materials is strongly influenced by the domain structure inherent to the compounds and the manner through which domain walls move in response to an applied field. In the absence of an applied field, domain walls form at pinning sites in the
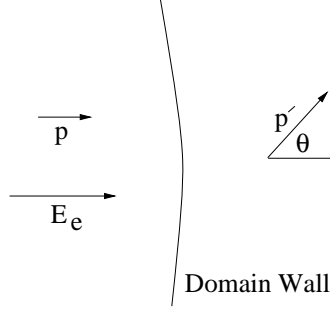
10

Figure 4. Domain wall and orientation of dipole moments $\mathbf{p}, \mathbf{p}'$ and electric field $\mathbf{E}_e$.

material (e.g., material inclusions or stress nonhomogeneities) to minimize the associated potential energy. Various experimental investigations have illustrated that at low input field levels, domain wall movement is reversible and can, at least conceptually, be attributed to the bending of domain walls [8, 18, 31]. For higher input fields, the local energy barriers associated with pinning sites are overcome and domain walls move for extended distances [8]. This translation of domain walls across pinning sites provides an irreversible mechanism contributing to the hysteresis observed in ferroelectric materials.

To quantify the resulting changes in the reversible polarization P_{rev}, irreversible polarization P_{irr}, and total polarization $P = P_{irr} + P_{rev}$, electrostatic energy principles are employed to quantify the energy required to reorient dipoles in the presence of an applied field. We summarize here the model developed in [44, 48, 49] and refer the reader to these references for details concerning the model.

We consider first the irreversible polarization P_{irr}. As detailed in [44], P_{irr} is quantified by determining the energy required to reorient a dipole in the presence of an effective field E_e as depicted in Figure 4. By employing the energy relation (20), the required change in energy is given by

$$\Delta \mathcal{E} = -\mathbf{p} \cdot \mathbf{E}_e + \mathbf{p}' \cdot \mathbf{E}_e \, .$$

The assumption that the energy $\mathcal{E}_{pin}$ required to break pinning sites is proportional to this change, along with integration over a control volume to obtain the change in polarization, yields the expression

$$\mathcal{E}_{pin}(P) = -k \int_0^P dP \tag{38}$$

for the energy required to break pinning sites. The parameter k is defined by

$$k = \frac{n \langle \mathcal{E}_\pi \rangle}{2p} \tag{39}$$

where n denotes the average density of pinning sites, $\langle \mathcal{E}_\pi \rangle$ is the average energy for 180° walls and p is an average dipole moment. Because the density and energy of individual pinning sites are unknown, the parameter k must be estimated for a given material. As detailed in [48], the parameter k is asymptotically related to the coercive field E_c for soft materials. Utilization of this property yields initial values for optimization routines and can aid in the adaptation of parameters to accommodate changing operating conditions.

The irreversible polarization is then quantified by determining the work required to achieve anhysteretic polarization levels minus the losses due to pinning. This yields the relation

$$\int_0^{E_e} P_{irr}(E_e) \, dE_e = \int_0^{E_e} P_{an}(E_e) \, dE_e - \int_0^{E_e} k \frac{dP_{irr}}{dE_e} \, dE_e \, . \tag{40}$$

11

Differentiation with respect to the effective field

$$E_e = E + \alpha P_{irr} \tag{41}$$

yields the local relation

$$P_{irr} = P_{an} - k\frac{dP_{irr}}{dE_e} \, .$$

Reformulation and quantification of the field orientation and reversal points yields the corresponding differential equation

$$\frac{dP_{irr}}{dE} = \widetilde{\delta}\frac{P_{an} - P_{irr}}{k\delta - \alpha\left(P_{an} - P_{irr}\right)} \, . \tag{42}$$

The anhysteretic polarization is specified by (35) with the effective field taken to be (41). The parameter $\delta = \text{sign}(dE)$ ensures that the energy required to break pinning sites always opposes changes in polarization while

$$\widetilde{\delta} = \left\{ \begin{array}{ll} 1 \, , & \{dE > 0 \text{ and } P < P_{an}\} \text{ or } \{dE < 0 \text{ and } P > P_{an}\} \\ 0 \, , & \text{otherwise} \, . \end{array} \right. \tag{43}$$

enforces that solely reversible polarization changes occur until the anhysteretic curve is reached.

The second component of the polarization is the reversible polarization which models the effects of domain wall bending. To first approximation, this is modeled by the relation

$$P_{rev} = c(P_{an} - P_{irr}) \tag{44}$$

where c is a parameter which must be estimated for the specific application (see [44]).

The total polarization is then given by

$$P = P_{rev} + P_{irr}$$

or equivalently,

$$P = cP_{an} + (1 - c)P_{irr} \, . \tag{45}$$

To implement the model, the effective field for a given field and irreversible polarization level is computed using (41). This effective field value is then employed in one of the expressions (35) to compute the corresponding anhysteretic polarization. The subsequent irreversible polarization is determined by numerically integrating (42). The total polarization is then specified by (45).

3.2 Ferromagnetic Compounds

The corresponding model for ferromagnetic materials, which was developed by Jiles and Atherton, preceded the ferroelectric model and motivated aspects of its development. The use of this model for general magnetic materials is detailed in [27, 28] whereas its use for characterizing magnetostrictive transducers is documented in [7, 11, 12, 13, 42]

In a manner analogous to ferroelectric materials, we let M_{rev} and M_{irr} respectively denote the reversible and irreversible components to the magnetization due to domain wall bending and translation. As detailed in [28], the use of magnetostatic principles to compute the energy required to reorient dipoles in order to model domain wall motion yields the differential equation

$$\frac{dM_{irr}}{dH} = \widehat{\delta}\frac{M_{an} - M_{irr}}{k\delta - \alpha(M_{an} - M_{irr})} \, . \tag{46}$$

The constant k, which is defined in a manner analogous to (39), quantifies irreversible effects. The anhysteretic magnetization is specified by (36) with the effective field given by

$$H_e = H + \alpha M_{irr} \quad \text{or} \quad H_e = H + \alpha M.$$

The parameter $\delta = \text{sign}(dH)$ again guarantees that the energy required to break pinning sites always opposes the field and $\tilde{\delta}$ (see (43) with E and P respectively replaced by H and M) enforces reversible domain wall motion until the anhysteretic curve is reached.

Reversible effects are modeled by employing magnetostatic energy relations to quantify the surface energy and corresponding forces required to bend domain walls. As detailed in [28], this yields the algebraic relation

$$M_{rev} = c(M_{an} - M_{irr}). \tag{47}$$

The summation of the irreversible and reversible components of the magnetization then yields the expression

$$M = (1 - c)M_{irr} + cM_{an} \tag{48}$$

for the total magnetization generated by an input field H in ferromagnetic materials.

3.3 Ferroelastic Materials

Domain wall movement in ferroelastic materials can be attributed to transitions in the domain structure between martensite variants or transitions between martensite and austenite phases [40]. The quantification of domain wall losses through the mechanisms described in Sections 3.1 and 3.2 for ferroelectric and ferromagnetic materials, has been employed for LiCsSO$_4$ by Tuszyński et al. [55] by employing a Landau-based anhysteretic model which accommodates second-order phase transitions. We summarize here the methodology for incorporating domain wall losses for general ferroelastic materials exhibiting either first-order (e.g., SMA's) or second-order phase transitions.

The characterization of the pinning energy is treated in an analogous manner to that for ferroelectric and ferromagnetic compounds. However, the quantification of the elastic energy required to reorient variants, and hence domains, is still under development so the ferroelastic loss mechanism should at this point be considered as phenomenological in nature. Details regarding the energy basis for this component will be presented in future work.

Letting ε denote the strain exhibited by the material, ε_{irr} denote the irreversible component to the strain, and ε_{an} denote the anhysteretic strain which would occur in the absence of defects, the energy required to reorient domains is assumed to have the form

$$\mathcal{E}_{pin}(\varepsilon) = k \int_0^{\varepsilon} d\varepsilon. \tag{49}$$

The constant k is assumed to be proportional to the average density of pinning sites (see (39)) and hence it provides a measure of the degree to which irreversible losses due to translation across energy minima will occur. Since the energy required to achieve a specified deformation state is comprised of the energy required to achieve thermodynamic equilibrium (the anhysteretic curve) minus the energy required to break pinning sites during lattice reorientation, the balance of elastic energy yields the relation

$$\int \varepsilon_{irr} d\sigma_e = \int \varepsilon_{an}(\sigma_e) d\sigma_e - k \int \frac{d\varepsilon_{irr}}{d\sigma_e} d\sigma_e. \tag{50}$$

Differentiation with respect to the effective stress yields a local loss relation. To provide a global model for quantifying the material behavior, it is necessary to account for the orientation of input

stresses which, for the 1-D model, is accomplished by defining $\delta = \text{sign}(\sigma_e)$. Reformulation of (50) then yields the differential equation

$$\frac{d\varepsilon_{irr}}{d\sigma} = \widetilde{\delta} \frac{\varepsilon_{an} - \varepsilon_{irr}}{k\delta - \alpha \left(\varepsilon_{an} - \varepsilon_{irr}\right)} \ . \tag{51}$$

The anhysteretic strain ε_{an} is determined by solving (37) with the effective stress $\sigma_e = \sigma + \alpha\varepsilon$. Details regarding the numerical algorithms employed for determining the roots of (37) will be provided in [47].

In a manner analogous to that employed for ferroelectric and ferromagnetic materials, reversible strains can be quantified through the algebraic expression

$$\varepsilon_{rev} = c(\varepsilon_{an} - \varepsilon_{irr})$$

which yields the model

$$\varepsilon = c\varepsilon_{an} + (1-c)\varepsilon_{irr} \tag{52}$$

for the total strains. We note that initial use of the model for characterizing SMA has revealed that for the considered samples, the reversible component to the strain appears negligible. In such cases, one can employ $c = 0$ so that the strain ε is directly specified by the differential equation (51).

3.4   General Ferroic Model

A comparison of the models for the polarization, magnetization and strains in ferroelectric, ferromagnetic and ferroelastic materials reveals that they exhibit a common structure. To formulate a general domain wall model for ferroic materials, we again employ the order parameters $e = P, M$ or ε and let $\widetilde{\varphi} = E, H$ or σ denote the conjugate fields. The three components of the model can then be specified in the following manner.

Irreversible:

$$\frac{de_{irr}}{d\widetilde{\varphi}} = \widetilde{\delta} \frac{e_{an} - e_{irr}}{k\delta - \alpha \left(e_{an} - e_{irr}\right)}$$

Reversible:

$$e_{rev} = c(e_{an} - e_{irr})$$

Total:

$$e = e_{rev} + e_{irr} = ce_{an} + (1-c)e_{irr}.$$

The anhysteretic components for ferroelectric, ferromagnetic and ferroelastic materials are respectively defined in (35), (36) and (37). We note that the differences in constitutive behavior for the individual materials are accommodated through the choice of free energy functionals and resulting anhysteretic relations summarized in Section 2.

4   Model Validation

The model developed in Section 3 provides a unified methodology for quantifying the hysteresis and constitutive nonlinearities inherent to ferroic materials under certain operating conditions. In this section, we present examples illustrating the performance of the model for characterizing piezoceramic, magnetostrictive and shape memory alloys.
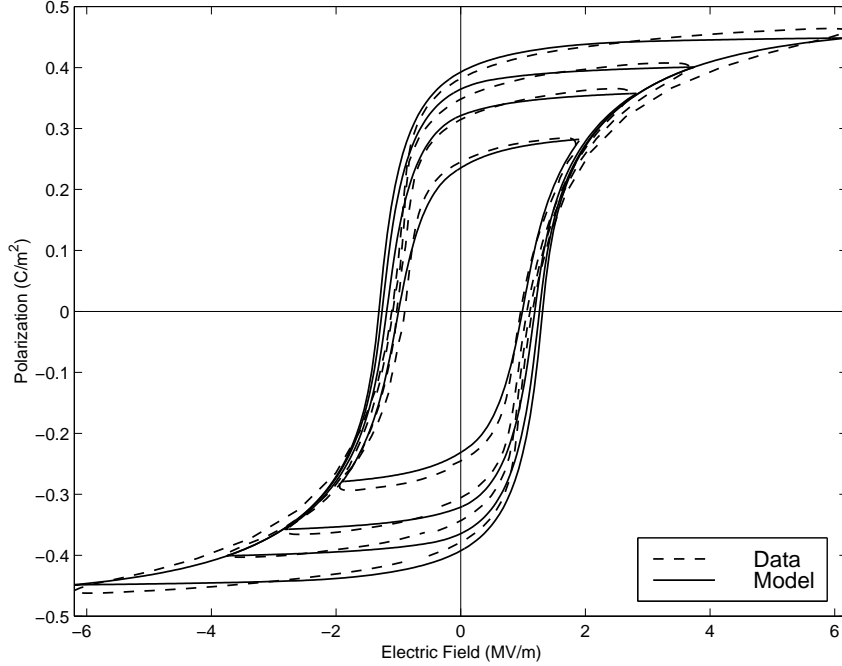
Figure 5. Model fit to 1600 V data for the circular PZT5A patch and model predictions at 600 V, 800 V and 1000 V with the parameter choices $\alpha = 3.6 \times 10^6$ Vm/C, $a = 4.2 \times 10^5$ V/m, $c = 0.30$, $k = 1.8 \times 10^6$ V/m and $P_s = 0.49$ C/m^2 (from [48]).

4.1 Ferroelectric Materials

The polarization model (45) has been employed to characterize the constitutive nonlinearities and hysteresis inherent to PZT5A, PZT5H, PZT4 [48] as well as the relaxor ferroelectric PMN-PT-BT operating in low-temperature, ferroelectric regimes [44]. Modifications of the model have also been developed to incorporate low frequency relaxation mechanisms for PZT [49] and the temperature-dependent transition between the ferroelectric, superparaelectric, and paraelectric states in relaxor ferroelectric materials [45, 46]. In concert, these investigations have illustrated the capability of this modeling strategy for a large number of ferroelectric and relaxor ferroelectric materials under a variety of operating conditions.

To illustrate the manner through which the model quantifies the polarization as a function of differing input field levels, we consider the characterization of a circular PZT wafer having a diameter of 2.54 cm and thickness of 0.0254 cm (10 mils). The data collected at 600 V, 800 V, 1000 V and 1600 V input levels is compared with models predictions in Figure 5. To obtain the parameters α, a, c, k and P_s, a least squares fit to the 1600 V data was performed to yield the parameter values $\alpha = 3.6 \times 10^6$ Vm/C, $a = 4.2 \times 10^5$ V/m, $c = 0.30$, $k = 1.8 \times 10^6$ V/m and $P_s = 0.49$ C/m^2. The model with these values was then used to predict the material response at the specified input levels. In all cases, the Langevin model (36) was used to quantify the anhysteretic component to the model. A comparison of the model solution with the experimental data illustrates that at all drive levels, the model is accurately predicting the E-P behavior of the physical material. This flexibility with regard to drive levels is provided in part by the energy formulation of the model. Additional examples illustrating the performance of the model and its extensions can be found in the previously cited references.
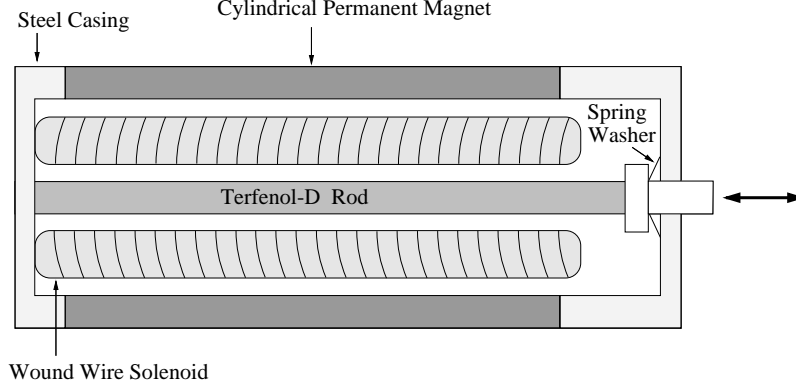
Figure 6. Cross section of a prototypical Terfenol-D magnetostrictive transducer (from [7]).

4.2 Ferromagnetic Materials

The magnetization model (48) is significantly more mature and has been tested under a wider variety of regimes than its polarization counterpart. The performance of the model for characterizing a variety of magnetic materials is illustrated in [27, 28] while the extension and validation for magnetostrictive transducers is detailed in [7, 11, 12, 13, 42]. We summarize here an example from [7] to illustrate aspects of the model.

Consider the prototypical magnetostrictive transducer depicted in Figure 6. A current $I(t)$ to the solenoid generates a magnetic field $H(t)$ which in turn causes dipoles in the central Terfenol-D rod to rotate. The spring mechanism maintains the rod in compression and further aligns dipoles perpendicular to the longitudinal rod axis in the absence of an applied field. The surrounding magnet provides the bias necessary to achieve bidirectional strains and can be used for flux shaping to minimize end effects in the rod. This transducer is typical of the construction employed in both academic and industrial applications.
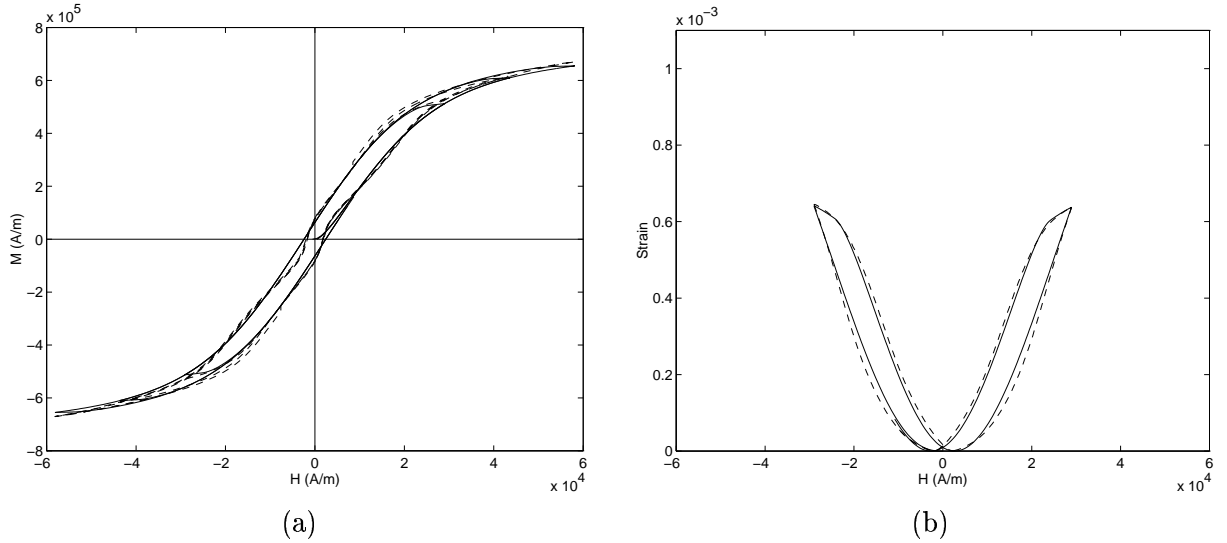

Figure 7. (a) Experimental data (– – –) and modeled relation (——) between the input field H and generated magnetization M; (b) Relation between input field H and generated strain e (from [7]).

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Under the experimental conditions detailed in [7], field, magnetization and strain measurements were collected for a variety of field and prestress levels. For a 1.0 ksi prestress, the H-M data colleted at two input levels is compared with the model solution in Figure 7a while measured and modeled lowfield strains are plotted as a function of field in Figure 7b. In the latter case, the quadratic relation $\lambda = \gamma M^2$ from (25) was used to model the strains. For the magnetization and strain levels illustrated here, the model accurately quantifies the nonlinear and hysteretic behavior of the transducer material. The extension of the model to utilize the full constitutive relation (25) is illustrated in [13] while the full coupling inherent to the magnetostrictive effect is addressed in [12].

4.3 Ferroelastic Materials

As a final example, we numerically illustrate the behavior of the anhysteretic and full strain model for ferroelastic compounds. Specifically, we consider the characterization of the shape memory alloy NiTi subjected to periodic tensile stresses with the temperature fixed at $T = 295$ K. In this regime, NiTi exhibits pseudoelastic behavior as illustrated by the experimental data of Shaw and Kyriakides [41].

The anhysteretic strains provided by the model (37) are compared with the total strains resulting from (52) in Figure 8. The parameters for the anhysteretic model are $\alpha_1 = 2.0810 \times 10^4$, $\alpha_2 = 9.5445 \times 10^6, \alpha_3 = 1.8630 \times 10^9, \alpha = -2.5414 \times 10^3$ while the coefficient k quantifying the degree of irreversible domain wall losses was taken to be $k = 2.8356$. It is observed that the incorporation of domain wall losses has the effect of smoothing the transition between the observed states. This is in accordance with observations made regarding the pseudoelastic behavior of a variety of shape memory compounds and general ferroic materials. The qualitative aspects of the model can be compared with experimental data in [41] that was collected under commensurate conditions. Note that in plotting the model solution, we have treated the input stress as the independent variable and generated strains as the dependent variable. The experimental validation of this component of the model is under investigation.
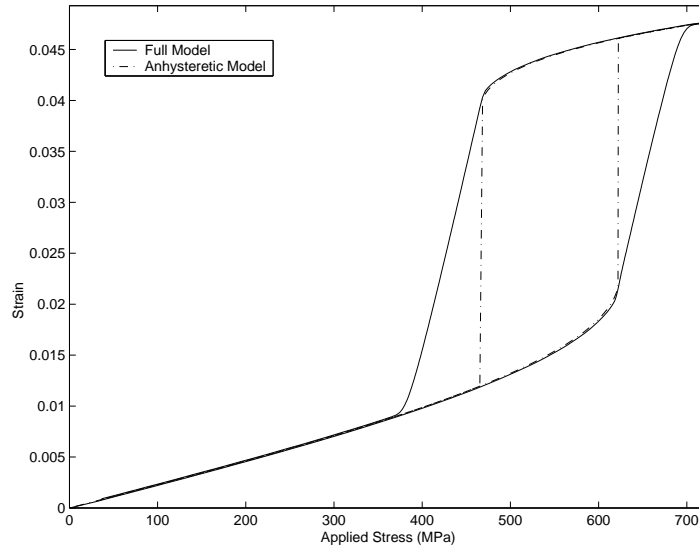

Figure 8. Response of the anhysteretic model (37) and full strain model (52) to a periodic tensile stress.

5 Concluding Remarks

The methodology presented here provides a unified technique for modeling hysteresis in ferroelectric, ferromagnetic and ferroelastic compounds. Hence it is applicable for piezoceramic, relaxor ferroelectric, magnetostrictive and shape memory alloys in quasistatic drive regimes. Extensions to the theory to accommodate phenomena such as relaxation effects in piezoceramic materials, eddy current losses in magnetostrictives, and temperature-dependencies in relaxor ferroelectrics are cited in the references.

The theory is constructed in two steps. In the first, thermodynamic principles are employed to quantify the anhysteretic response of the materials in the absence of pinning sites which restrict domain wall motion. This quantifies aspects of domain switching in the materials. Because the microscopic mechanisms which produce domain switching differ between ferroelectric, ferromagnetic and ferroelastic materials, this component of the model is the most phenomenological. To quantify the effects of domain wall bending and translation, the electrostatic, magnetostatic, and elastic energy required to reorient dipoles or elastic variants (e.g., martensite variants) is considered in conjunction with macroscopic averaging of material properties to obtain quasi-macroscopic models having a small number of effective parameters. As illustrated in the examples and cited references, the physical nature of the models facilitates the estimation of parameters from easily measured characteristics of the data. Asymptotic relations between the parameters and characteristics of the data can also be employed to update parameters to accommodate changes in operating conditions. Finally, full and inverse compensators based on the model have been employed for initial control design in high performance systems utilizing smart material actuators operating in nonlinear and hysteretic regimes [35, 43, 50].

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